

# Atomic parity nonconservation in $\text{Ra}^+$

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We report on a theoretical analysis of the suitability of the  $7s^2S_{1/2} \leftrightarrow 6d^2D_{3/2}$  transition in singly ionized radium to measure parity nonconservation, in the light of an experiment planned at the KVI of the University of Groningen. Relativistic coupled-cluster theory has been employed to perform an *ab initio* calculation of the parity nonconserving electric dipole amplitude of this transition, including single, double, and leading triple excitations. We discuss the prospects for a sub-1% precision test of the electroweak theory of particle physics.

In atomic systems, parity is broken due to the exchange of the neutral vector boson  $Z^0$ , which mediates the weak interaction between the atomic electrons and the quarks in the nucleus. This atomic parity nonconservation (APNC) gives rise to small parity nonconserving electric dipole transition amplitudes ( $E1_{\text{PNC}}$ ). The APNC effect gets strongly enhanced in heavy atoms and can be measured by the interference of  $E1_{\text{PNC}}$  with a suppressed electromagnetic transition amplitude ( $M1$ ,  $E2$ ) [1, 2]. The accurate measurement of the  $6s^2S_{1/2} \leftrightarrow 7s^2S_{1/2}$  transition in atomic  $^{133}\text{Cs}$  by the Boulder group [3, 4] constitutes a precision test of the electroweak sector of the Standard Model (SM) of particle physics [5]. By combining the measurement with a many-body atomic structure calculation, the weak nuclear charge could be determined [6].

The importance of APNC to particle physics is a strong incentive to further pursue these challenging experiments. With the experimental and theoretical accuracies at an impressive 0.35% [3, 4] and 0.5% [6] level, respectively, the  $^{133}\text{Cs}$  result agrees with the SM prediction within one standard deviation. Nevertheless, it is desirable to consider other candidates for APNC studies, see *e.g.* Ref. [7]. New experiments have been proposed for Cs [8] and Fr [9] atoms. Of special interest is the proposal by Fortson to measure APNC in *one single* laser-cooled and trapped ion [2]. Such single-ion experiments offer important benefits, such as long coherence times and precise control of various systematic effects. Promising ions from the experimental and atomic-theory point of view are heavy alkali-like ions, in particular  $\text{Ba}^+$  and  $\text{Ra}^+$  [10]. Proof-of-principle experiments have been carried out with  $^{138}\text{Ba}^+$  by the Fortson group [10, 11, 12].

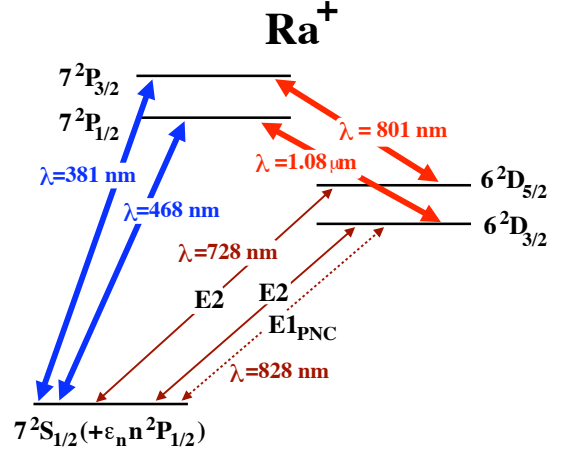


FIG. 1: Relevant energy levels in  $\text{Ra}^+$ .

At the TRI $\mu$ P facility [13, 14] at the accelerator institute KVI in Groningen an APNC experiment on  $\text{Ra}^+$  is in progress [15]. An important advantage of  $\text{Ra}^+$  is that all relevant transitions are in the optical regime, *cf.* Fig. 1, and thus are accessible by commercially available solid-state laser technology. The goal is to measure the  $E1_{\text{PNC}}$  amplitude of the  $7s^2S_{1/2} \leftrightarrow 6d^2D_{3/2}$  transition. We address here the question what the prospects are to push the corresponding atomic theory below 1%, such that the experiment can serve as a high-precision test of the SM. We analyze various relevant properties of  $\text{Ra}^+$  and assess the remaining uncertainties.

The parity-nonconserving nuclear-spin independent (NSI) interaction is due to the electron-quark neutral weak interaction, the Hamiltonian of which is given by

$$H_{\text{PNC}}^{\text{NSI}} = \frac{G_F}{2\sqrt{2}} Q_W \gamma_5 \varrho_{\text{nuc}}(r), \quad (1)$$

where  $G_F$  is the Fermi constant,  $\varrho_{\text{nuc}}$  the nuclear density,

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and  $\gamma_5$  is the standard Dirac matrix;  $Q_W$  is the weak nuclear charge, which is equal to  $(2Z + N)c_{1u} + (2N + Z)c_{1d}$  in terms of the coupling constants of the electron to the up and down quarks;  $Z$  and  $N$  are the number of protons and neutrons. The Hamiltonian in Eq. (1) mixes atomic states of opposite parity but with the same angular momentum. Its strength is weak enough to consider it as a first-order perturbation. We therefore write the valence state ( $v$ ) atomic wave function as

$$|\Psi_v\rangle = |\Psi_v^{(0)}\rangle + G_F |\Psi_v^{(1)}\rangle, \quad (2)$$

where  $|\Psi_v^{(0)}\rangle$  is the atomic wave function of the Dirac-Coulomb (DC) Hamiltonian ( $H_{DC}$ ) and  $|\Psi_v^{(1)}\rangle$  is the first-order correction due to the PNC NSI interaction.

To a first-order approximation, the  $E1_{\text{PNC}}$  transition amplitude between the  $7s\ ^2S_{1/2}$  ( $= i$ ) and  $6d\ ^2D_{3/2}$  ( $= f$ ) states is given by

$$E1_{\text{PNC}} = G_F \frac{\langle \Psi_f^{(0)} | D | \Psi_i^{(1)} \rangle + \langle \Psi_f^{(1)} | D | \Psi_i^{(0)} \rangle}{\sqrt{\langle \Psi_f^{(0)} | \Psi_f^{(0)} \rangle \langle \Psi_i^{(0)} | \Psi_i^{(0)} \rangle}}, \quad (3)$$

which, after expansion, takes the form

$$E1_{\text{PNC}} = \sum_{I \neq i} \frac{\langle \Psi_f^{(0)} | D | \Psi_I^{(0)} \rangle \langle \Psi_I^{(0)} | H_{\text{PNC}}^{\text{NSI}} | \Psi_i^{(0)} \rangle}{(E_i - E_I) \sqrt{\langle \Psi_f^{(0)} | \Psi_f^{(0)} \rangle \langle \Psi_i^{(0)} | \Psi_i^{(0)} \rangle}} + \sum_{J \neq f} \frac{\langle \Psi_f^{(0)} | H_{\text{PNC}}^{\text{NSI}} | \Psi_J^{(0)} \rangle \langle \Psi_J^{(0)} | D | \Psi_i^{(0)} \rangle}{(E_f - E_J) \sqrt{\langle \Psi_f^{(0)} | \Psi_f^{(0)} \rangle \langle \Psi_i^{(0)} | \Psi_i^{(0)} \rangle}}, \quad (4)$$

where  $D$  is the electric dipole ( $E1$ ) operator,  $I$  and  $J$  represent the allowed intermediate states, and  $E$  is the energy of the state. An accurate determination of  $E1_{\text{PNC}}$  depends on the precision of the matrix elements of  $D$  and of  $H_{\text{PNC}}^{\text{NSI}}$ , and of the energy differences between the different states. At the same time, it is also important to take all intermediate states into account, something which is not possible in the often-used sum-over-states approach. We therefore employ the relativistic coupled-cluster (RCC) theory, which allows us to evaluate the properties to all orders in perturbation theory. The RCC method was previously used to calculate APNC in  $^{137}\text{Ba}^+$  with sub-1% accuracy [16].

We obtain the first-order wave functions of Eq. (3) in the RCC framework as the solution of

$$G_F (H_{DC} - E_v) |\Psi_v^{(1)}\rangle = -H_{\text{PNC}}^{\text{NSI}} |\Psi_v^{(0)}\rangle, \quad (5)$$

where  $v$  stands for valence electron, which is either  $i$  or  $f$ . The unperturbed and perturbed wave functions are expressed as

$$|\Psi_v^{(0)}\rangle = \exp(T^{(0)}) \{1 + S_v^{(0)}\} |\Phi_v\rangle, \quad (6)$$

and

$$\begin{aligned} |\Psi_v^{(1)}\rangle &= \exp(T') \{1 + S_v'\} |\Phi_v\rangle \\ &= \exp(T^{(0)}) (T^{(1)} \{1 + S_v^{(0)}\} + \{S_v^{(1)}\}) |\Phi_v\rangle, \end{aligned} \quad (7)$$

respectively, where  $|\Phi_v\rangle$  is the mean-field wave function determined with the Dirac-Fock (DF) method.  $T$  and  $S_v$  are the core and valence-core RCC correlation operators, respectively, where the superscript 0 indicates in the presence of the Coulomb interaction, the prime ( $'$ ) indicates in the presence of both the Coulomb and APNC interaction, and 1 indicates their linear approximations. Substituting the above expressions in Eq. (3), we obtain

$$E1_{\text{PNC}} = G_F \frac{\langle \Phi_f | C_f^\dagger \overline{D^{(0)}} C_i | \Phi_i \rangle}{\sqrt{(1 + N_f^{(0)})(1 + N_i^{(0)})}}, \quad (8)$$

where

$$\begin{aligned} N_v^{(0)} &= \langle \Phi_v | S_v^{(0)\dagger} \exp(T^{(0)\dagger}) \exp(T^{(0)}) S_v^{(0)} | \Phi_v \rangle, \\ C_v &= T^{(1)} \{1 + S_v^{(0)}\} + S_v^{(1)}, \end{aligned} \quad (9)$$

and  $\overline{D^{(0)}} = \exp(T^{(0)\dagger}) D \exp(T^{(0)})$ . The matrix element is evaluated using the generalized Wick's theorem [16].

Our RCC work has two salient features. We evaluate  $E1_{\text{PNC}}$  by using the direct solution of the first-order perturbed equation as given in Eq. (5) rather than summing over a finite number of intermediate states [17]. The core correlation effects modified by the parity nonconserving weak interaction are evaluated to all orders through  $T^{(1)}$  in the framework of the relativistic CCSD(T) method. These effects cancel strongly in Cs and Fr, where both the initial and final states are  $S$ -states. However, it is essential to consider them accurately in the  $S$ - $D$  transitions in  $\text{Ba}^+$  and  $\text{Ra}^+$ , where these contributions are significant. For our calculation, we have used numerical DF/ $V^{N-1}$  orbitals to describe the occupied and bound virtual orbitals. The continuum states were represented by  $V^{N-1}$  Gaussian-type orbitals (GTOs) [18] using the parameters  $\alpha = 5.25 \times 10^{-3}$  and  $\beta = 2.73$ . The finite size of the nucleus is accounted for by assuming a Fermi charge distribution [18].

In Table I, we present our RCC results for the  $E1_{\text{PNC}}$  amplitude of the  $7s\ ^2S_{1/2} \leftrightarrow 6d\ ^2D_{3/2}$  transition in the isotope  $^{226}\text{Ra}^+$ . Shown are the results of the DF method, of the RCC method with single and double excitations (CCSD), and with the leading triple excitations (CCSD(T)). The difference between the CCSD(T) and CCSD results is small. Our best value is the CCSD(T) result  $E1_{\text{PNC}} = 46.4 \times 10^{-11} \text{iea}_0 (-Q_W/N)$ . Also shown are two results of Dzuba *et al.* [19]: in a sum-over-states

TABLE I:  $E1_{\text{PNC}}$  for the  $7s\ ^2S_{1/2} \leftrightarrow 6d\ ^2D_{3/2}$  transition in the isotope  $^{226}\text{Ra}^+$ , in units of  $10^{-11} \text{iea}_0 (-Q_W/N)$ .

	<i>This work</i>	Ref. [19]
DF	40.4	
CCSD	46.1	Mixed-states 42.9
CCSD(T)	46.4	Sum-over-states 45.9

approach they found  $45.9 \times 10^{-11} iea_0(-Q_W/N)$ ; in a mixed-states approach, wherein the APNC interaction explicitly mixes states of opposite parity, they obtained  $42.9 \times 10^{-11} iea_0(-Q_W/N)$ . Neither calculation includes structural radiation, the weak correlation potential, and normalization of states, effects which are included by us.

The  $E1_{\text{PNC}}$  amplitude for the  $6s\ ^2S_{1/2} \leftrightarrow 7s\ ^2S_{1/2}$  transition in Cs is about  $0.9 \times 10^{-11} iea_0(-Q_W/N)$  [6]. Thus, the APNC effect in  $\text{Ra}^+$  is larger by a factor close to 50. In heavy atoms, ANPC gets enhanced by the overall factor  $K_r Z^2 Q_W(Z, N)$ , where  $Q_W \sim N \sim Z$ , and  $K_r$  is a relativistic factor that depends on the nuclear charge and radius. This is the “faster-than- $Z^3$  law” [1], which implies that  $\text{Ra}^+$  is favored over Cs by a factor of about 20. An additional factor of around 2 can be understood as follows. For Cs (and Fr) the  $S$ - $S$  transition is used, for  $\text{Ra}^+$  (and  $\text{Ba}^+$ ) the  $S$ - $D$  transition. Since the  $Z^0$ -boson is very heavy, the weak interaction between electrons and the quarks in the nucleus has (almost) zero range. The overlap of the electrons with the nucleus is largest for the  $S$  states, and thus the mixing of the  $P$ -states into the  $S$  states gives the major contribution to  $E1_{\text{PNC}}$ . However, in Cs and Fr the initial and final  $S$  states contribute with opposite signs, which leads to a significant cancellation in  $E1_{\text{PNC}}$ . In fact, for Cs there are three dominant terms in the sum over the states, which add up to a total value that is half the size of the largest individual term [6].

The  $S$ - $D$  transitions in  $\text{Ba}^+$  and  $\text{Ra}^+$  do not suffer from such a cancellation, since the contribution from the  $D$ -state to APNC is small. In Table II, we analyze which intermediate states contribute most to the total sum. Clearly, in contrast to the Cs  $S$ - $S$  case, the sum is strongly dominated by one term: the contribution from the  $7p\ ^2P_{1/2}$  state. These qualitative results are robust, they are consistent with the findings of Ref. [19], and they are, in fact, already borne out by a simple calculation with quantum-defect theory, analogous to Ref. [20]. This simple estimate gives for Cs, Fr,  $\text{Ba}^+$ , and  $\text{Ra}^+$  results accurate to some 10%; for  $\text{Ra}^+$  we find  $E1_{\text{PNC}} = 45(4) \times 10^{-11} iea_0(-Q_W/N)$ .

In Table III, we present our results for the excitation energies,  $E1$  transition amplitudes, and hyperfine constants for the relevant transitions and states in  $\text{Ra}^+$ . We also list experimental values where available. For the excitation energies, we compare to the only available spec-

TABLE II: The contributions to the  $E1_{\text{PNC}}$  from the different  $P$ -states (%).

State	type	%	State	type	%
$6p\ ^2P_{1/2}$	core	8.7	$8p\ ^2P_{1/2}$	bound	-3.3
$6p\ ^2P_{3/2}$	core	-15	$9p\ ^2P_{1/2}$	bound	-0.7
$7p\ ^2P_{1/2}$	bound	111	$10p\ ^2P_{1/2}$	continuum	-0.1
$7p\ ^2P_{3/2}$	bound	-2.6	$11p\ ^2P_{1/2}$	continuum	1.1

TABLE III: Excitation energies,  $E1$  transition amplitudes, and  $A_I/g_I$  for different low-lying states of  $\text{Ra}^+$ .

Transition	$7s\ ^2S_{1/2}$ $7p\ ^2P_{1/2}$	$7s\ ^2S_{1/2}$ $7p\ ^2P_{3/2}$	$6d\ ^2D_{3/2}$ $7p\ ^2P_{1/2}$	$6d\ ^2D_{3/2}$ $7p\ ^2P_{3/2}$
<i>Excitation energy</i> [ $\text{cm}^{-1}$ ]				
This work	21509	26440	9734	14665
Experiment [21]	21351	26209	9267	14125
<i>E1 transition amplitude</i> [a.u.]				
This work	3.31	4.58	3.68	1.56
Ref. [19]	3.223	4.477	3.363	1.504
GTOs [22]	3.28	4.54	3.64	1.54
State	$7s\ ^2S_{1/2}$	$7p\ ^2P_{1/2}$	$7p\ ^2P_{3/2}$	$6d\ ^2D_{3/2}$
<i>Hyperfine interaction constant</i> ( $A_I/g_I$ ) [MHz]				
This work	19689.37	3713.75	312.91	441.67
Experiment [23]	18772	3691	314.12	-

troscopy measurement [21], which dates back to 1933. For the  $E1$  transition amplitudes, for which there are no experimental data, we list the results of Ref. [19] and of our previous work [22] using GTOs. Therein, the lifetimes of the metastable  $D$ -states were calculated to be  $0.627(4)$  s for  $6d\ ^2D_{3/2}$  and  $0.297(4)$  s for  $6d\ ^2D_{5/2}$ .

Since the hyperfine structure is a good probe of the wave functions at the nucleus, we have, in order to estimate the accuracy of the  $H_{\text{PNC}}^{\text{NSI}}$  matrix elements, calculated the ratio between the magnetic dipole hyperfine structure ( $A$ ) and the nuclear gyromagnetic ( $g$ ) constants, neglecting isotope effects, and compared these with experimental results for  $\text{Ra}^+$  from ISOLDE [23, 24]. Our calculated value for  $[A_I/g_I(7S_{1/2})A_I/g_I(7P_{1/2})]^{1/2}$  differs by 3% from the experimental value, which is a reasonable estimate for the dominant uncertainty in the atomic theory. Thus, our best value for the parity nonconserving  $E1$  amplitude in  $\text{Ra}^+$  is  $E1_{\text{PNC}} = 46.4(1.4) \times 10^{-11} iea_0(-Q_W/N)$ .

It appears feasible to push the accuracy of the atomic theory for  $\text{Ra}^+$  to the sub-1% level. Improvements along several lines are in progress. The Breit interaction [25, 26] and QED corrections, which contribute around 1%, need to be included. The neutron-skin effect [27], which also contributes at the sub-1% level, has to be investigated. However, at the same time it is clear that experimental information to test the atomic theory is severely lacking. Not all relevant energy levels are known [21], there is no experimental information on the  $E1$  transition strengths, nor on the lifetimes of the  $D$ -states. It is highly desirable to have more experimental data on the magnetic dipole and electric quadrupole hyperfine constants  $A$  and  $B$  for the various  $\text{Ra}^+$  isotopes. The extraction of these constants [23, 24] is model dependent, and ideally one would like to use a single consistent *ab initio* framework for this.

TABLE IV: The properties of the isotopes of  $\text{Ra}^+$  suitable for a single-ion APNC experiment.  $A$  is the mass number,  $I$  is the nuclear spin, and  $\tau_{1/2}$  the half-life time.

$A$	$I$	$\tau_{1/2}$	Possible production reaction
213	$1/2^-$	2.74(6) min	$^{208}\text{Pb} + ^{12}\text{C} \rightarrow ^{213}\text{Ra} + 7\text{n}$
223	$3/2^+$	11.43(5) d	$\text{p} + ^{232}\text{Th} \rightarrow ^{223}\text{Ra} + ^A\text{X} + \text{an} + \text{bp}$
224	$0^+$	3.6319(23) d	$\text{p} + ^{232}\text{Th} \rightarrow ^{224}\text{Ra} + ^A\text{X} + \text{an} + \text{bp}$
225	$1/2^+$	14.9(2) d	$^{229}\text{Th} \rightarrow ^{225}\text{Ra} + \alpha$
226	$0^+$	1600(7) y	Commercially available
227	$3/2^+$	42.2(5) min	$\text{p} + ^{232}\text{Th} \rightarrow ^{227}\text{Ra} + ^A\text{X} + \text{an} + \text{bp}$

At the TRI $\mu$ P facility, radium isotopes can be produced in fusion and evaporation or spallation reactions. The ions can be collected in a radio-frequency trap where they can be laser-cooled on the  $7s\ ^2S_{1/2} \leftrightarrow 7p\ ^2P_{1/2}$  resonance line at 468 nm, with repumping via the  $6d\ ^2D_{3/2} \leftrightarrow 7p\ ^2P_{1/2}$  transition at 1.08  $\mu\text{m}$ , for which strong lasers are available. They will then be transferred to a miniature trap for the single-ion experiment, where techniques similar to Refs. [10, 11, 12] will be applied to perform the measurements. In particular, the  $7s\ ^2S_{1/2} \leftrightarrow 7p\ ^2P_{3/2}$  and  $6d\ ^2D_{5/2} \leftrightarrow 7p\ ^2P_{3/2}$  transitions at 381 nm and 801 nm, respectively, can be used for “shelving” [10].

A list of the  $\text{Ra}^+$  isotopes suitable for a single-ion experiment is shown in Table IV. A half-life of the order of seconds is required for a high-precision single-ion experiment, but, on the other hand, it should not exceed a few days, so as to avoid long-lived radioactive contamination of the core equipment. Good candidates therefore are the odd isotopes  $^{213}\text{Ra}^+$  and  $^{227}\text{Ra}^+$ , and the even isotope  $^{224}\text{Ra}^+$ . (Since the odd isotopes have a nonzero nuclear spin, the nuclear spin-dependent weak interaction will contribute to  $E1_{\text{PNC}}$  [28]). The isotopes listed can all be produced at TRI $\mu$ P, and the experimental data required to constrain the atomic theory can be measured there with laser spectroscopy. Since multiple  $\text{Ra}^+$  isotopes will be available, the possibility exists to measure APNC in a chain of isotopes, which can help to eliminate remaining uncertainties in the atomic theory [29].

In conclusion,  $\text{Ra}^+$  appears to be an excellent candidate for an APNC experiment, since  $E1_{\text{PNC}}$  is large, the required lasers are all at convenient wavelengths, and one can exploit the high-precision techniques of single-ion trapping. The atomic theory needed for the interpretation of the experiment could reach an accuracy better than 1%, but precise experimental data for the relevant atomic properties are mandatory to achieve such a benchmark. The prospects for APNC in  $\text{Ra}^+$  as a precise test of the SM look promising.

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